Journal of Hazardous Materials 168 (2009) 145–155

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

A means to estimate thermal and kinetic parameters of coal dust layer from hot surface ignition tests

Haejun Park, Ali S. Rangwala*, Nicholas A. Dembsey

Department of Fire Protection Engineering, Worcester Polytechnic Institute, 100 Institute Road, Worcester, MA 01609, United States

ARTICLE INFO

Article history:
Received 9 July 2008
Received in revised form 31 January 2009
Accepted 3 February 2009
Available online 12 February 2009

Keywords:
Coal dust
Hot-surface
Ignition
Property estimation
Spontaneous combustion

ABSTRACT

A method to estimate thermal and kinetic parameters of Pittsburgh seam coal subject to thermal runaway is presented using the standard ASTM E 2021 hot surface ignition test apparatus. Parameters include thermal conductivity (k), activation energy (E), coupled term (QA) of heat of reaction (Q) and pre-exponential factor (A) which are required, but rarely known input values to determine the thermal runaway propensity of a dust material. Four different dust layer thicknesses: 6.4, 12.7, 19.1 and 25.4 mm, are tested, and among them, a single steady state dust layer temperature profile of 12.7 mm thick dust layer is used to estimate k, E and QA. k is calculated by equating heat flux from the hot surface layer and heat loss rate on the boundary assuming negligible heat generation in the coal dust layer at a low hot surface temperature. E and QA are calculated by optimizing a numerically estimated steady state dust layer temperature distribution to the experimentally obtained temperature profile of a 12.7 mm thick dust layer. Two unknowns, E and QA, are reduced to one from the correlation of E and QA obtained at criticality of thermal runaway. The estimated k is 0.1 W/m K matching the previously reported value. E ranges from 61.7 to 83.1 kJ/mol, and the corresponding QA ranges from 1.7×10^9 to 4.8×10^{11} J/kg s. The mean values of E (72.4 kJ/mol) and QA (2.8×10^{10} J/kg s) are used to predict the critical hot surface temperatures for other thicknesses, and good agreement is observed between measured and experimental values. Also, the estimated E and QA ranges match the corresponding ranges calculated from the multiple tests method and values reported in previous research.

1. Introduction

Thermal runaway, also described as supercritical self-heating or spontaneous ignition, has been considered as a serious hazard in many industrial processes and applications such as bulk coal stockpiles [1–3], nickel–cadmium accumulators [4], and dust material deposits on a heated surface [5]. Ignition in dust deposits by thermal runaway can also lead to subsequent dust explosions [6].

Various methods and techniques have been developed to evaluate the propensity of thermal runaway of a material: hot plate test, oven-basket test, thermal analysis test, etc. [7]. Since thermal runaway can occur in various circumstances, each test method has its own merits in application. The hot plate test is specifically designed to evaluate the thermal runaway hazard of a granular material up to a couple of centimeters, a realistic thickness in many industrial environments. The bottom surface of the dust layer is exposed to a hot plate while the top surface is cooled in ambient air. Despite being a relatively short and easy test procedure and resembling actual hazardous conditions closer than the other test methods in terms of sample amount and configuration, hot plate test has been considered just an approximate screening method on the basis of 'go/no go' criteria [8,9]. The oven-basket test, historically the most common test method, represents thermal runaway of granular materials surrounded by constant temperature. This method requires a wire mesh basket, usually cubical in shape, which contains the test material to be placed in an oven at a high temperature. Due to small sample dimensions compared to a real storage size, a high oven temperature is required to cause thermal runaway. The test results are then extrapolated to assess the hazard of a realistic storage stockpile. A review of the oven test and its application to a realistic scenario is given by Jones [10,11]. Thermal analysis tests such as thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) can measure the critical decomposition temperatures and heat energy produced by the chemical reactions. In these two tests, heat transfer phenomena are of less concern relative to chemical reaction [12] due to a tiny amount of test sample.

Parameters required for determining the thermal runaway hazard of a dust material including thermal conductivity, total heat transfer coefficient, activation energy, and pre-exponential factor in an Arrhenius equation are rarely known or are test environment

* Corresponding author. Tel.: +1 858 344 1946.
E-mail address: rangwala@wpi.edu (A.S. Rangwala).

0304-3894/$ – see front matter © 2009 Elsevier B.V. All rights reserved.
doi:10.1016/j.jhazmat.2009.02.010

specific. Some of these values can be obtained from other test methods such as TGA and DSC, and multiple hot plate tests with different dust layer thicknesses. The objectives of this study are to estimate these parameters from the hot plate test with a single thickness as compared to the multiple tests method by optimizing a numerical solution to match the experimental temperature distribution in the dust layer. Pittsburgh seam coal dust, one of the benchmark test materials of ASTM E 2021 [13], is used as the test material.

2. Background

The thermal runaway theory by Semenov [14] assumes constant temperature distribution throughout a reaction zone with heat loss at the boundary. However, Semenov's theory is only applicable to cases such as a well-stirred gas mixture due to the assumption of no thermal resistance in the reaction zone. Frank-Kamenetskii [15] adopted temperature distribution in the reaction zone, but assumed no heat loss on the boundary. Limitations of each of these cases were overcome by Thomas and Bowes [18] taking into account temperature distribution both in a reaction zone and heat loss on the boundaries [16].

Thermal runaway implies a sudden temperature increase due to thermal imbalance between heat generation rate and loss rate. Heat generation rate is known to follow the Arrhenius equation as an exponential function of temperature, and heat loss rate can be represented as a linear function of temperature as shown in Fig. 1(A). For an asymmetrically heated dust layer, heat transferred from the hot surface increases the dust layer temperature and consequently leads to a higher heat generation rate. Heat generated by exothermic reaction at the elevated temperature in the layer competes with the heat loss by convection and radiation at the top surface. Therefore, for a given material, layer thickness, and ambient environment, the hot surface temperature is the variable which determines the occurrence of either a thermal balance or thermal runaway. In Fig. 1(A), if the hot plate temperature (T_p) is set and remains at T_{p3} where heat generation rate in the dust layer is equal to the loss rate at the boundary, dust layer temperature will remain in steady state. However, any slight increase of heat generation rate can lead to thermal runaway. If T_p is set and remains lower than T_{p3} such as at T_{p2} where heat generation rate in the dust layer is higher than heat loss rate, the dust layer temperature will increase up to the point A where a thermal balance exists. A perturbation can cause temperature to increase beyond the point A, but the higher heat loss rate between T_{p2} and T_{p3} will direct it back to the steady state point A. Another thermal balance point C can be seen to be unstable since below point C heat loss rate is higher than heat generation rate, which yields temperature drop, and above point C thermal runaway occurs. In case of heating process with starting temperature lower than T_{p3}, point A is the only thermal balance point. As hot surface temperature increases from T_{p2} to T_{p3}, point C decreases and merges with point A resulting in point B. If T_p is set and remains at T_{p4} which is just above T_{p3}, dust layer temperature continuously increases and reaches thermal runaway.

Presumed temperature distributions in an asymmetrically heated dust layer with thickness $2r$ are shown in Fig. 1(B). A dust layer comes into contact with a hot surface at $x = 0$. Line 1 represents linear temperature distribution of an inert material without internal heat generation. The slope is determined by the heat loss rate on the top boundary at $x = 2r$ and thermal conductivity with a given T_p at T_{p1}. Curve 2 represents steady state condition at a low hot plate temperature with relatively small amount of heat generation in the dust layer. Curve 3 represents the maximum steady state condition of a dust layer. The maximum layer temperature (T_m) is observed at x_m very close to the hot surface. Curve 4 represents a transient temperature profile of thermal runaway. Higher oxygen concentration near the open boundary causes more reaction, and consequently, higher temperature than the lower area of the dust layer. The layer ignition temperature (LIT) or the minimum hot plate temperature for thermal runaway exists at T_{p4}. This is the main concern in most cases, and can be derived from the analytical solution of T_{p3}, since thermal runaway is theoretically expected to occur just above T_{p3}.

The analytical solution for T_{p3}, the maximum hot plate temperature for the dust layer to remain in steady state, is available from Thomas's thermal runaway model [17] with the assumption of negligible reactant depletion [18,19]. Assuming the dust layer as an infinite slab, a one dimensional steady state heat conduction equation can be written as,

$$k\frac{\partial^2 T}{\partial x^2} = -\rho Q A e^{-E/RT}, \tag{1}$$

where k = thermal conductivity of dust layer (W/m K), T = temperature (K), ρ = density (kg/m^3), Q = heat of reaction (J/kg), and A = Arrhenius pre-exponential factor (1/s). Q and A can be treated as one combined term (QA) for mathematical convenience. In the exponential part, E = the activation energy (J/mol) and R = the universal gas constant (=8.314 J/mol K).

Boundary conditions of constant temperature at the bottom surface and Newtonian cooling on the top surface are,

$$T = T_p \quad \text{at } x = 0, \tag{2a}$$

$$-k\frac{dT}{dx} = h_t(T_s - T_a) \quad \text{at } x = 2r, \tag{2b}$$

where $h_t = h_c + h_r$ = total heat transfer coefficient (W/m^2 K) accounting for convective (h_c) and radiant (h_r) heat transfer. T_s = top surface temperature (K), T_a = ambient temperature (K).

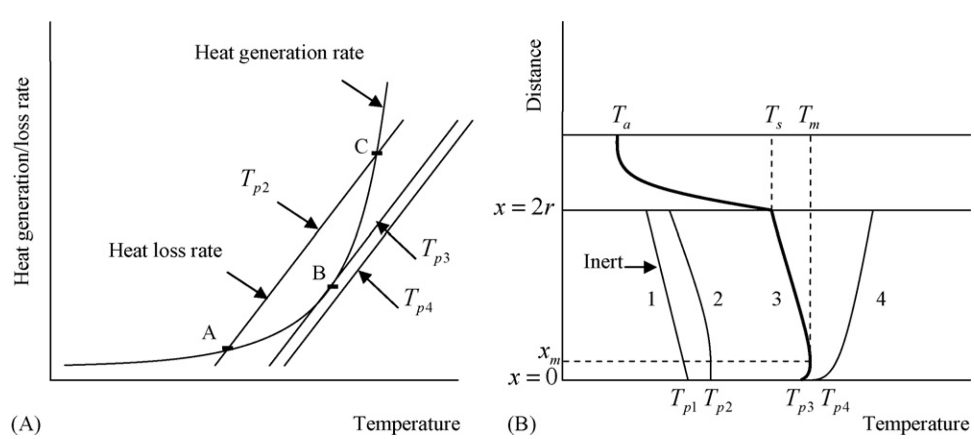

Fig. 1. Thermal runaway concept (A) and temperature distributions in asymmetrically heated dust layer (B).